

Current-Induced Step Bending Instability on Vicinal Surfaces

Da-Jiang Liu¹, John D. Weeks^{1,2}, and Daniel Kandel³

¹*Institute for Physical Science and Technology, University of Maryland, College Park, Maryland 20742*

²*Department of Chemistry, University of Maryland, College Park, Maryland 20742*

³*Department of Physics of Complex System, Weizmann Institute of Science, Rehovot 76100, Israel*

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We model an apparent instability seen in recent experiments on current induced step bunching on Si(111) surfaces using a generalized 2D BCF model, where adatoms have a diffusion bias parallel to the step edges and there is an attachment barrier at the step edge. We find a new linear instability with novel step patterns. Monte Carlo simulations on a solid-on-solid model are used to study the instability beyond the linear regime.

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Vicinal crystal surfaces can exhibit a number of different morphological instabilities that may be important in crystal growth and device fabrication. One of the most interesting of these instabilities arises from the biased diffusion of adatoms under the influence of an external driving force, as illustrated by the step bunching instability on Si(111) surfaces [1] induced by heating with a direct electric current. Many different aspects of this so-called electromigration problem have been investigated when the driving force is normal to the steps: e.g., the microscopic origin of the driving force on adatoms, the effect of temperature on the instability, and the resulting step patterns [2–5]. We show here that when the relative orientation of the external driving force and the miscut angle of the vicinal surface is varied, there exists a qualitatively different instability that can be explored both experimentally and theoretically.

Because of the intrinsic anisotropy of a stepped surface, kinetic instabilities can usually be associated with one of the two principal axes normal to and along the average step orientation (the x and y directions, respectively). It is convenient to describe step positions in Fourier space. From a linear stability analysis, the evolution of small perturbations in the positions of the steps away from a uniform step train $\delta x_n(y, t)$ can be written as

$$\delta x_n(y, t) = \sum_{q, \phi} \delta x(q, \phi) e^{iqy + in\phi + \omega(q, \phi)t} + \text{c.c.}, \quad (1)$$

where q is a wavenumber along the step direction, and ϕ a phase factor relating consecutive steps. When $\text{Re } \omega(q, \phi) > 0$, a perturbation of that mode will grow exponentially.

One class of instabilities involves the growth of finite q perturbations along the y (step) direction, as illustrated by the Bales-Zangwill [6] instability. In this example it is sufficient to take $\phi = 0$, since this (in-phase) mode is maximally unstable and will dominate. Indeed the underlying (Mullins-Sekerka) instability can be understood by considering a single interface.

Another class of instabilities involves the relative motion of neighboring steps, and can usually be understood

using a one dimensional (1D) model with $q = 0$ and the step pairing mode $\phi = \pi$. Examples include the step bunching instability due to either the Ehrlich-Schwoebel barrier [7,8], impurity adsorption [9–11], or the usual electromigration instability with the current normal to the steps. Stoyanov [2] used a 1D model to study the latter instability in detail. He showed that a vicinal surface can be unstable towards step bunching when adatoms have a diffusion bias in the step-down direction and there exists an extra barrier for attachment to step edges. The step pairing mode ($\phi = \pi$) gives the maximum instability unless there are very strong repulsions between steps [12] or significant direct interterrace diffusion [13,14].

Quantitative studies of electromigration based on a generalized 1D step model have shown very good agreement with many aspects of the experiments [13]. However, a more general perspective is required to understand some recent surprising results by Latyshev *et al.* [15]. They observed the sudden appearance of *step bends* in certain regions of the surface where the step edges are nearly parallel to the electric current, leading to the formation of *anti-step* (down-step) bunches as well as the usual (up-) step bunches. Motivated by this result, we study here a 2D step model with biased diffusion *parallel* to the step edge direction, and indeed find an instability. This new instability requires *both* finite q and finite ϕ and reaches its maximum when the phase shift $\phi = \pm\pi/2$ at small q , unlike any other surface instability we are aware of.

Figure 1 gives a schematic view of the surface and introduces the notation. As in Stoyanov's model [2], adatoms have a drift velocity $D_s F/kT$, where D_s is the adatom diffusion rate on flat terraces. F is the magnitude of the external force acting on adatoms, and we assume in this case it is in the y direction. For simplicity, here we ignore evaporation and deposition of adatoms on terraces; this does not affect the basic instability we find. We make the usual quasistatic approximation [16], setting $\partial c/\partial t = 0$ in the diffusion equation describing the adatom concentration field $c(x, y)$ on the terraces:

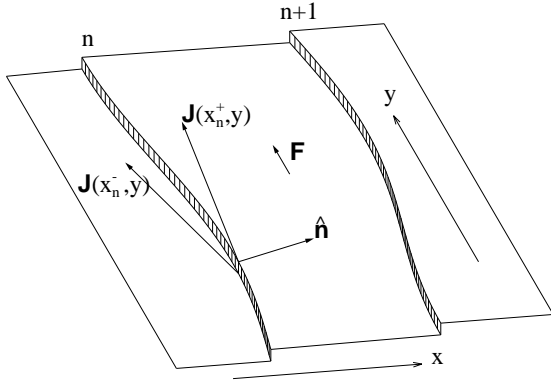


FIG. 1. Schematic view of a vicinal surface with an external force parallel to the step edge.

$$D_s \left(\frac{\partial^2 c}{\partial x^2} + \frac{\partial^2 c}{\partial y^2} \right) - \frac{D_s F}{kT} \frac{\partial c}{\partial y} = 0. \quad (2)$$

The boundary conditions are given by linear kinetic expressions with a parameter κ governing the rate of exchange of atoms between steps and terraces:

$$\pm \kappa [c_n^{\text{eq}}(y) - c(x_n^\pm(y), y)] = \hat{\mathbf{n}} \cdot \mathbf{J}(x_n^\pm(y), y), \quad (3)$$

where the surface adatom flux \mathbf{J} is given by

$$\mathbf{J}(x, y) = -D_s (-\partial/\partial x, -\partial/\partial y + f) c(x, y), \quad (4)$$

and $f \equiv F/kT$. The $+$ ($-$) indicates the ascending (descending) terrace next to step n as shown in Fig. 1. $c_n^{\text{eq}}(y)$ is the local equilibrium adatom concentration of step n at position y , and $\hat{\mathbf{n}}$ is the unit vector normal to the step edge. Through microscopic mass conservation, the normal velocity of step n can be written as

$$\begin{aligned} v_n &= a^2 \hat{\mathbf{n}} \cdot [\mathbf{J}(x_n^+, y) - \mathbf{J}(x_n^-, y)] \\ &= D_s a^2 [2c_n^{\text{eq}}(y) - c(x_n^+, y) - c(x_n^-, y)], \end{aligned} \quad (5)$$

where a is the lattice constant in the x - y plane, assuming a square lattice.

To take into account the stabilizing effects of the line tension and step repulsions, we write

$$c_n^{\text{eq}}(y) = c_0^{\text{eq}} \exp \left(\frac{\mu_n(y) - \mu_C}{kT} \right), \quad (6)$$

where the step edge chemical potential $\mu_n(y)$ — the change in free energy per atom for adding atoms to the step at position y — is given by [17,18]

$$\mu_n = a^2 [V'(w_n) - V'(w_{n-1}) + \tilde{\beta} \partial^2 x_n / \partial y^2] + \mu_C. \quad (7)$$

Here we assume there is a nearest-neighbor repulsive interaction $V(w)$ between steps separated by a terrace width w and $V'(w) = dV/dw$. $\tilde{\beta}$ is the step edge stiffness and μ_C is the equilibrium atom chemical potential of the crystal.

To carry out a linear stability analysis [19], we substitute the solution of the diffusion equation Eq. (2) with boundary conditions Eq. (3) into Eq. (1). Keeping terms linear in δx , we find for the growth rate $\omega(q, \phi)$:

$$\omega(q, \phi) = \frac{-2D_s c_{\text{eq}}^0 a^2 \Lambda_q [f q d \sin \phi + g(q, \phi)]}{2\Lambda_q d \cosh(\Lambda_q w_0) + (\Lambda_q^2 d^2 + 1) \sinh(\Lambda_q w_0)}, \quad (8)$$

where

$$\begin{aligned} g(q, \phi) &= g_x (1 - 2 \cos \phi + \cos 2\phi) - g_y q^2 \cos \phi \\ &\quad + [g_y q^2 + 2g_x (1 - \cos \phi)] \\ &\quad \times [\cosh(\Lambda_q w_0) + \Lambda_q d \sinh(\Lambda_q w_0)], \end{aligned} \quad (9)$$

$$\Lambda_q = \sqrt{q^2 + i f q}, \quad (10)$$

and

$$g_x = \frac{a^2}{kT} \frac{d^2 V}{dw^2} \Big|_{w=w_0}; \quad g_y = \frac{\tilde{\beta} a^2}{kT}. \quad (11)$$

In the above equations there is an important length scale $d \equiv D_s/\kappa$, the ratio between the adatom diffusion rate and the attachment rate, as introduced by Pimpinelli *et al.* [20]. $g(q, \phi)$ is the stabilizing term from the line tension and step repulsions; it reduces to the known result of Pimpinelli *et al.* [20] in the case when $g_x = 0$ (i.e., ignoring step repulsions) and $f \rightarrow 0$.

The first term in the square bracket of Eq. (8) is the major result of this paper. For $f > 0$ (external force in the $+y$ direction), $\text{Re} \omega$ is positive for $-\pi < \phi < 0$ and is maximally unstable when $\phi = -\pi/2$ for a particular wavenumber q that is not too large [for large enough q , the relaxation term $g(q, \phi)$ will dominate]. This is quite different from the usual step pairing instability with maximum instability at $\phi = \pi$ if the bias is in the step down direction. Figure 2 is an illustration of a step train with $\phi = -\pi/2$. For several neighboring steps, the hills and troughs are shifted in their positions and form regions of high step densities (narrow terraces) that extend along certain direction. Changing the sign of f (the direction of the external force) makes modes with $0 < \phi < \pi$ unstable. Note that if the attachment rate is very fast (so that $\kappa \rightarrow \infty$ or $d \rightarrow 0$), the mass transport is limited by the diffusion rate and the instability disappears. Thus the extra barrier for adatom attachment to step edges is crucial for the existence of the instability.

We can gain a better understanding of the physical origin of this instability from the following qualitative picture. Figure 3 draws three steps with perturbations in the form of sinusoids with a phase shift $\phi = -\pi/2$ (steps are out-of-phase with every other step). We assume there is a very strong barrier for adatom attachment to a step edge. The direction of the atomic flux

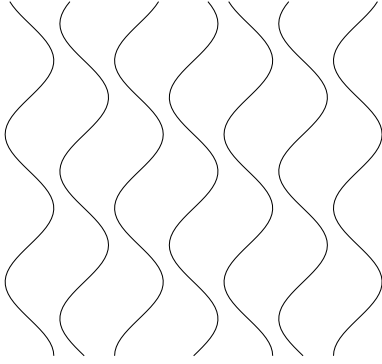


FIG. 2. Illustration of a step train with sinusoidal fluctuations with phase shift $-\phi = \pi/2$.

on a terrace driven by an external force parallel to the average step edge direction is then approximately the average of the directions of the two steps bounding the terrace.

From microscopic mass conservation [Eq. (5)], the velocity of a segment of a step is given by the difference of the normal components of the two surface atomic fluxes at the two sides of the step. For segments A, C, and E in Fig. 3, the two fluxes cancel and the velocity vanishes at those points. However for segments B and D, there is a net contribution from the two fluxes that makes the step move in the direction shown. Thus the amplitude of the perturbation will increase and the step train is unstable. Changing the direction of the external force will reverse the direction of the motion and stabilize the perturbation of Fig. 3, but will destabilize a perturbation with phase shift $\phi = \pi/2$. Thus a vicinal surface is unstable when there is driven diffusion *parallel* to the step edge direction. This picture is only correct when there are no efficient channels for adatoms to hop directly from one terrace to another one.

It is natural and straightforward to extend the linear stability analysis to an external force in a general direction. The general solution is too complicated to write down here, but in the limit of weak external forces and small wavenumber q , we have

$$\omega \approx \frac{2D_s c_{eq}^0 a^2}{2d + w_0} \left[-2f_x d \frac{1 - \cos \phi}{2d + w_0} - f_y q d \sin \phi - g(q, \phi) \right] \quad (12)$$

where $f_{x(y)} = F_{x(y)}/kT$ is the $x(y)$ component of the (reduced) external force. The term proportional to f_x has the familiar $(1 - \cos \phi)$ form for the pairing instability for an external force normal to the steps.

These results may help us understand the origin of the anti-step bunches seen by Latyshev *et al.* [15] on Si(111) at elevated temperatures. Initially the current direction is *perpendicular* to the average step direction and step bunching occurs in the usual way. Because of sublima-

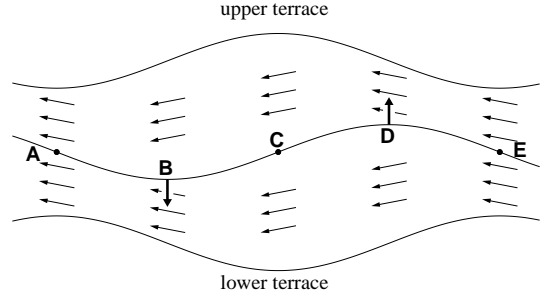


FIG. 3. Illustration of the atomic flux induced by an external field parallel to the average step edge direction on two terraces bounded by three steps with sinusoidal perturbations and phase shift $\phi = -\pi/2$.

tion, nearly uniform arrays of *crossing steps* form between the step bunches [4]. As the system coarsens and the distance between bunches gets larger, the inclination angle of the crossing steps increases and becomes nearly perpendicular to the step bunches, and thus parallel to the current direction. It is at this stage that Latyshev *et al.* [15] observe the appearance of *step bends* in the crossing arrays that grow in size and accumulate into an anti-step bunch, i.e., a step bunch in the opposite direction to the initial orientation of the vicinal surface.

Since the physical model discussed in this paper also give good agreement with the initial step bunching behavior of Si(111) [13] when the current is normal to the steps, it seems quite likely that the instability discussed above is responsible for the appearance of the step bends in the crossing arrays. However, a detailed analysis of the anti-step bunch formation in the experiments necessarily involves the nonlinear regime and a treatment of interactions with the preexisting step bunches, which is beyond the scope of this letter.

To get some information about the nonlinear behavior, we carried out kinetic Monte Carlo simulations of a solid-on-solid (SOS) model [13] that incorporates the essential physical features of biased diffusion with attachment barriers at steps. The external force is mimicked by an asymmetry in the attempt frequencies for exchange of surface atoms between nearest-neighbor sites. The energy barrier is simply the binding energy of the surface atom in its initial site, except that there is an *extra* barrier when the movement causes a change in total energy, and hence for attachment at steps (see Ref. [13] for more details). Figure 4 is a snapshot of a simulation with the driving force parallel to the average step edge direction. The initial step edges are in the vertical direction and adatoms have a downward diffusion bias. The dark regions are step bunches formed by step segments bending in the opposite direction to the individual steps (black lines) on the terrace. Note that only a relatively small region of each step lies within the step bunch, in contrast to the usual step bunches that form when the current

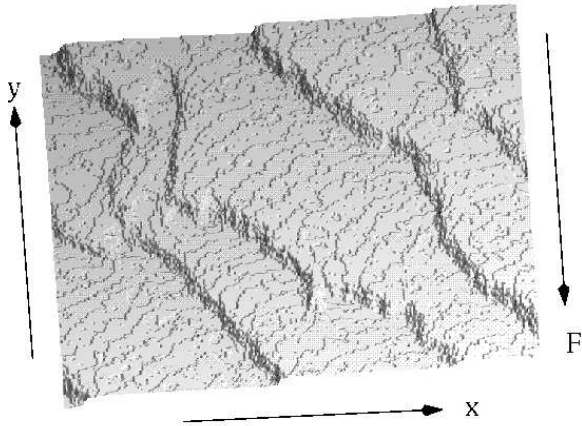


FIG. 4. Snapshot of a simulation using a solid-on-solid model with a diffusion bias parallel to the step edge direction. The initial steps are along the vertical (y) direction.

is normal to the step direction. This general feature is already suggested from the patterns in Fig. 2.

The earlier SOS simulations of Dobbs and Krug [22] gave very different results. They used Metropolis dynamics, which does not incorporate the step edge barrier, and the resulting step patterns contained several artifacts and did not compare well with experiments on Si(111).

Krug and Dobbs [21,22] also introduced a continuum model that showed that a surface is quite generally unstable under an external driving force when the adatom mobility has a slope dependence. This prediction, unlike other features, e.g., the orientation of selected facets and the coarsening rate, is independent of the underlying physical origin of the slope dependence (in our case, the step edge barrier) and is thus robust. Indeed one can make connection between the step model and the continuum model by taking $\phi \rightarrow q_x w_0$ and $q \rightarrow q_y$ and expanding Eq. (12) for small $q_{x(y)}$:

$$\omega \sim f_x q_x^2 + (2d + w_0) w_0^{-1} f_y q_x q_y + \mathcal{O}(q^4) \quad (13)$$

Except for an anisotropic ratio that is absent from their model, this equation is qualitatively the same as the result of the continuum model in Ref. [22] for a surface tilted away from a high symmetry plane with the broken symmetry of the underlying crystal structure along the x direction. Although their continuum model does not capture all the essential physics of a vicinal surface below the roughening temperature, e.g., the anisotropy in the adatom mobility and the singularity in surface stiffness at zero slope, it does give an idea of the instability at very large length scales.

In conclusion we report a new linear instability on vicinal surface that arises from the interplay between a step train and the two dimensional diffusion fields between the steps. There are many interesting theoretical issues yet to be resolved. The study of the instability beyond the linear regime is needed to understand the coarsening

behavior and anti-step bunch formation. The effect of a current along the step direction is also being investigated experimentally [23].

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